PATENT SPECIFICATION

DRAWINGS ATTACHED

Inventors: HISAO YAMAMOTO and NOBORU IWAASA

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COMPLETE SPECIFICATION

Synthesis Reaction Apparatus

We, UBE KOSAN KABUSHIKI KAISHA, a corporation organised under the laws of Japan, of 1 of No. 1976 Oaza Oguchi, Ube-Shi, Yamaguchi-Ken, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to an apparatus for performing exothermic catalytic synthesis reactions, such as ammonia synthesis and methanol synthesis, and which is equipped with means for temperature control of the catalyst bed.

Such a kind of synthesis reaction apparatus may ordinarily be classified into external heat exchange type and self heat exchange type according to the mode of removing the heat of reaction generated.

The external heat exchange type of apparatus is generally complicated in its construction due to the flow system of thermal medium which does not participate in the synthesis reaction. Moreover, it involves the disadvantage that the temperature of the catalyst bed may become too high upstream of the coolers while becoming too low downstream thereof.

The self heat exchange type of apparatus involves the problem of difficulty of effective temperature control of the catalyst bed for carrying out an isothermal operation.

Thus, with regard to the ammonia synthesis reaction, the reaction velocity decreases progressively with increase in concentration of the ammonia. This means that the heat generated per unit volume of catalyst decreases. Consequently, at the inlet portion of catalyst bed a large quantity of heat is generated which causes rapid elevation of the bed temperature, while, on the other hand, at the outlet portion a relatively small quantity of heat is generated which causes only gradual elevation of the temperature of that portion of the bed.

It is thus often expedient for carrying out an effective isothermal operation that the thermal conductivity of the catalyst bed should be greater in the vicinity of the inlet portion than in the vicinity of the outlet portion.

For this purpose, there has been proposed a method of passing unreacted cool gas concurrently with reaction gas and a method of passing the same countercurrently therewith.

However, in the concurrent system, there is a tendency for the catalyst bed temperature of the reaction gas outlet portion to become higher. From a consideration of the reaction velocity and the equilibrium concentration, it is obvious that a lower temperature for a higher concentration of ammonia is advantageous to the reaction. On the other hand, in the countercurrent system, the catalyst bed temperature of the reaction gas inlet portion can become too high, causing deterioration of the catalyst, while the bed temperature of the outlet portion can become so low that the reaction temperature is only with difficulty maintained.

According to the present invention, there is provided an apparatus for an exothermic synthesis reaction, comprising a cylindrical reaction chamber having longitudinally disposed therein a plurality of catalyst tubes communicating at one end with a heat exchanger within said chamber, means for supplying cool reaction gas to said heat exchanger, and temperature control means for delivering cool reaction gas in controllable amounts to portions of said catalyst tubes intermediate the ends thereof for heat exchange therewith, the arrangement being such

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that in operation of the apparatus reaction gas supplied to said heat exchanger is heated therein by exchange with hot reacted gas from said catalyst tubes and is then passed through said reaction chamber in contact with said catalyst tubes and is introduced into the other ends of said tubes for reaction therein, said cool reaction gas being delivered to said intermediate portions in amounts sufficient to maintain the reaction temperature in said catalyst tubes in the desired range.

For a better understanding of the invention and to show how the same may be carried into effect reference will now be made, by way of example, to the accompanying drawings, in which:—

Figure 1 is a longitudinal cross-sectional view of an ammonia synthesis reaction apparatus equipped with means for temperature control of catalyst bed;

Figure 2 is a transverse cross-sectional view of the apparatus shown in Figure 1 taken on the line A—A′ of Figure 1;

Figure 3 shows, on a larger scale, a fragment of the apparatus shown in Figure 1 (corresponding approximately to part B of Figure 1); and

Figure 4 shows a graph of temperature change along the catalyst bed of the apparatus of Figure 1.

Referring now to Figure 1 of the accompanying drawings, the apparatus comprises an inner cylinder 2 placed in an outer cylinder 1 so as to define therebetween an annular space 3. The top of the outer cylinder 1 is closed in gas-tight manner by a closure 4. An inlet 5 for cool unreacted gas is located at a position close to the top of the outer cylinder 1 and a main inlet 6 for unreacted gas is located at a position close to the bottom of cylinder 1. A spacer ring 8 having a number of perforations 7 is disposed in the annular space 3 adjacent the top of the inner cylinder 2. The inner cylinder 2 is provided in its upper portion with a reaction chamber 9 and, in its lower portion with a heat exchange chamber 10.

The inner cylinder 2 is closed in gas-tight manner at the top thereof by an insulation plate 11. The reaction chamber 9 and heat exchange chamber 10 are partitioned by a gas collection chamber 12 and the heat exchange chamber 10 and the inside of the bottom portion of the outer cylinder 1 are partitioned by a further gas collection chamber 13 which has an annular space around the circumference thereof.

The reaction chamber 9 is divided into a plurality of sections by a plurality of chambers 16 each formed by a pair of baffle plates 14 and 15 extending perpendicular to the axis of the inner cylinder 2. A plurality of catalyst tubes 17 mutually in parallel is longitudinally arranged in the reaction chamber 9. The catalyst tubes 17 are open at the tops thereof and communicate at the bottom with the gas collection chamber 12. The catalyst tubes 17 are constricted at the position of each chamber 16 so as to form small gaps 29 between the tubes and thereby allow communication of the chambers 16 with the inside of the inner cylinder 2. Each catalyst tube 17 is packed with catalyst 28.

An unreacted gas distribution nipple 18 is mounted at the bottom of the reaction chamber 9, to allow direct communication of the reaction chamber 9 with the heat exchange chamber 10 across the gas collection chamber 12. The reaction chamber 9 is further equipped centrally thereof with temperature control means T for the catalyst beds in the catalyst tubes 17.

The temperature control means T comprises a plurality of pipes 20 for cool gas disposed in a temperature control tube 19, the space between the pipes 20 and the inside wall of the temperature control tube 19 being filled with insulation material. The temperature control means T extends downwardly into the reaction chamber 9 through the closure 4 and the insulation plate 11. The inlet of each gas pipe 20 is connected to an outside transfer line (not shown in the drawings) and the outlet opens into a chamber 16, one pipe 20 being provided for each chamber 16.

The heat exchange chamber 10 is provided with a plurality of baffle plates 21 and a plurality of tubes 22 for reacted gas, which tubes 22 extend from the gas collection chamber 12 to the other gas collection chamber 13 through the heat exchange chamber 10. An annular slit 23 is formed by the gap between the baffle plate at the lower end of the cylinder 2 and the upper surface of the gas collection chamber 13, whereby the heat exchange chamber 10 is placed into direct communication with the inside of the outer cylinder 1.

A conduit 24 is provided for introducing preheated unreacted gas in the start-up operation of the reaction apparatus and for charging unreacted cooling gas in normal operation to prevent local heating of the top portion of the catalyst bed. Passages 25 and 26, which communicate with the conduit 24, are bored in the insulation plate 11 of the inner cylinder 2 for those purposes. An outlet 27 is provided for reacted gas.

The operation of the apparatus is as follows.

A major portion of unreacted gas enters the apparatus through the main inlet 6. Another portion of unreacted gas enters at the inlet 5 for unreacted cooling gas in order to cool the outer cylinder 1, passes through the perforations 7 of the spacer ring 8, and flows down through the annular space 3. The unreacted gas and the unreacted cooling gas mix and flow into the heat exchange cham-

ber 10 of the inner cylinder 2 by way of the suit 23 provided at the bottom of the heat exchange chamber 10. The mixture of unreacted gases is heated as it passes between the baffle plates 21 by heat exchange with the reacted gas flowing down through the tubes 22 and the heated mixture enters the reaction chamber 9 through the gas distribution nipple 18.

The unreacted gas flows upwardly through the gaps between the catalyst tubes 17 and between the outer face of the catalyst tubes 17 and the inner face of the reaction chamber 9 and cools the catalyst tubes 17.

At the same time, cool gas flows in the cooling gas pipes 20 of the temperature control means T from outside transfer lines (not shown in the drawings) and is fed into each chamber 16. Then this cool gas passes through gaps 29 formed between the constricted portions of the catalyst tubes 17, is dispersed into the reaction chamber 9, is uniformly mixed with the unreacted gas therein resulting in a reduction of the temperature thereof, and flows upwardly while cooling the catalyst tubes 17. The gas thus adjusted to a suitable temperature for the synthesis reaction enters the catalyst tubes 17 at the top thereof and is reacted while passing therethrough by the action of the catalyst 28 packed in the catalyst tubes 17.

The reacted gas containing about 20% of ammonia flows from the bottom of the catalyst tubes 17 into the gas collection chamber 12 and thence passes downwards in the tubes 22 in the heat exchange chamber 10. The reacted gas is heat exchanged while flowing down the tubes 22 with the unreacted gas which enters the heat exchange chamber 10 from the slit 23, and is discharged from the outlet 27 via the gas collection chamber 13 to the outside of the outer cylinder 1.

The useful heat of the discharged product gas is recovered in a waste heat boiler to generate about 0.8 ton of steam per ton of ammonia.

The temperature control means T installed in the apparatus prevents excessive cooling of the outlet portion of the catalyst bed by reducing the efficiency of heat exchange of the unreacted cool gas at the outlet portion of catalyst bed by decreasing the supply velocity thereof, and at the same time prevents the overheating of the inlet portion of catalyst bed by increasing the efficiency of heat exchange of the unreacted cool gas at the inlet portion of catalyst bed by increasing the supply velocity thereof. Further, the temperature control means T is effective in preventing the local heating of catalyst bed.

In general, the location most liable to exhibit violent reaction, i.e. the occurrence of rapid temperature elevation has been found to shift gradually towards the outlet of catalyst bed when deterioration of catalyst 28 occurs. In such cases, an appropriate utilization of the present temperature control means T can counteract successfully the undesirable change of temperature.

The following Example illustrates the invention.

EXAMPLE

A nitrogen-hydrogen mixture (ratio of gases of about 1:3 by volume) containing 1.8% by volume of ammonia was used as feed gas for an ammonia synthesis, reaction being carried out in an apparatus as described above with reference to the accompanying drawings. The temperature of the catalyst bed was maintained at from 450 to 520°F and the pressure at 300 atmospheres.

Unreacted gas entered through the main inlet 6 at a temperature of 150°C, was heated by exchange with the tubes 22 in which the reacted gas flowed at a temperature of from 330 to 450°C to raise the temperature of the unreacted gas to 390°C at the gas distribution nipple 18. The temperature of the gas finally became 490°C at the inlet of the catalyst tubes 17.

The temperature of unreacted gas employed for cooling was 25°C, this gas being introduced into the apparatus through the inlet 5, through the cool gas pipes 20 and through the unreacted cooling gas conduit 24. The unreacted gas was cooled from about 490° to about 420°C by means of the cool gas from the outlets of the pipes 20.

The outlet temperature of the catalyst tubes 17 was 450°C and the temperature in the reacted gas tube 22 was 330°C.

Figure 4 shows a curve indicating the temperature of the gas passing through the apparatus, the labelled parts of the curve corresponding to parts of the apparatus as follows:—

a corresponds to the gas at the point of entry through the inlet 6;

b corresponds to the region of the nipple 18;

c—d, e—f and g—h corresponds to the gas passing through the chambers 16 in the region of the constricted portions of the catalyst tubes 17;

i corresponds to the gas on entry at the top of the catalyst tubes;

j corresponds to the point of exit of the gas from the catalyst tubes; and

k corresponds to the gas at the outlet nozzle 27.

Thus, part a—i of the curve indicates the temperature of the unreacted gas and part i—k represents the temperature of the reacted gas.

WHAT WE CLAIM IS:—

1. An apparatus for an exothermic synthesis reaction, comprising a cylindrical reaction chamber having longitudinally disposed therein a plurality of catalyst tubes communi-

cating at one end with a heat exchanger within said chamber, means for supplying cool reaction gas to said heat exchanger, and temperature control means for delivering cool reaction gas in controllable amounts to portions of said catalyst tubes intermediate the ends thereof for heat exchange therewith, the arrangement being such that in operation of the apparatus reaction gas supplied to said heat exchanger is heated therein by exchange with hot reacted gas from said catalyst tubes and is then passed through said reaction chamber in contact with said catalyst tubes and is introduced into the other ends of said tubes for reaction therein, said cool reaction gas being delivered to said intermediate portions in amounts sufficient to maintain the reaction temperature in said catalyst tubes in the desired range.

2. An apparatus according to Claim 1, wherein said temperature control means comprises a longitudinal cylindrical member disposed centrally in said reaction chamber, said member containing a plurality of gas pipes each of which communicates with the interior of said reaction chamber in the vicinity of a portion of said catalyst tubes intermediate the ends thereof for supply of cool gas thereto.

3. An apparatus according to Claim 2, wherein said reaction chamber is provided with baffle plates enclosing said portions of said catalyst tubes, thereby to allow efficient contacting of said tubes by said cool gas.

4. An apparatus according to Claim 2 or 3, wherein said catalyst tubes are of reduced cross-section at said portions thereof.

5. An apparatus according to any preceding claim, wherein said heat exchanger is formed with a plurality of tubes for reacted gas communicating at one end with the ends of said catalyst tubes and at the other end with the exterior of the reaction chamber.

6. An apparatus according to Claim 5, wherein said heat exchanger is provided with a plurality of baffle plates to direct reaction gas around said tubes for reacted gas.

7. An apparatus according to any preceding claim, wherein said cylindrical reaction chamber is housed within an outer cylinder annularly spaced from said chamber, said annular space communicating at one end with a supplementary supply of cool reaction gas and at the other end with the heat exchanger, whereby supplementary cool reaction gas can be introduced into said annular space and be delivered therefrom into the heat exchanger.

8. An apparatus according to any preceding claim, wherein conduit means are provided which communicate with said reaction chamber in the vicinity of said other ends of said catalyst tubes for the introduction therein of preheated reaction gas during start-up and of cool reaction gas during operation to prevent over-heating of said other ends of said catalyst tubes.

9. An apparatus for an exothermic synthesis reaction, substantially as hereinbefore described with reference to, and as shown in, Figures 1,2 and 3 of the accompanying drawing.

HASELTINE, LAKE & CO,
Chartered Patent Agents,
28 Southampton Buildings, Chancery Lane,
London, W.C.2.
Agents for the Applicants.

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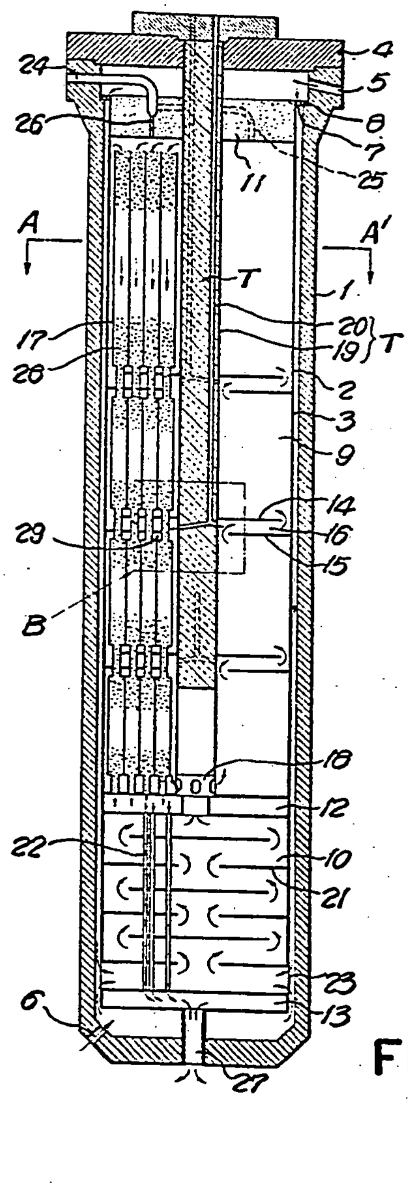
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FIG.1.

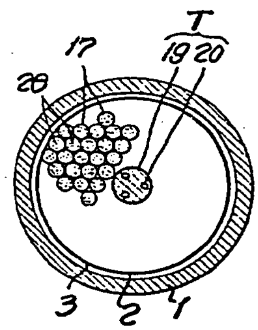
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FIG.2.

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2 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheets 1 & 2

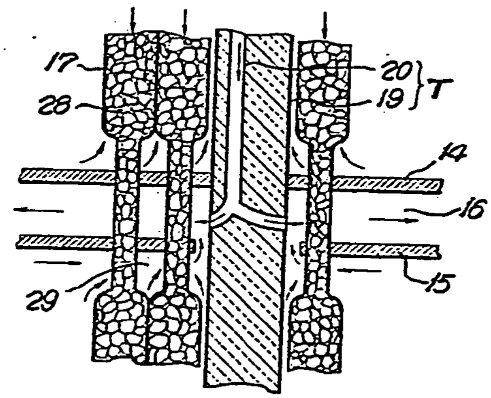
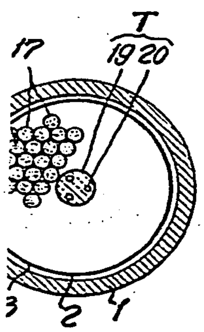


FIG.3.



G.2.

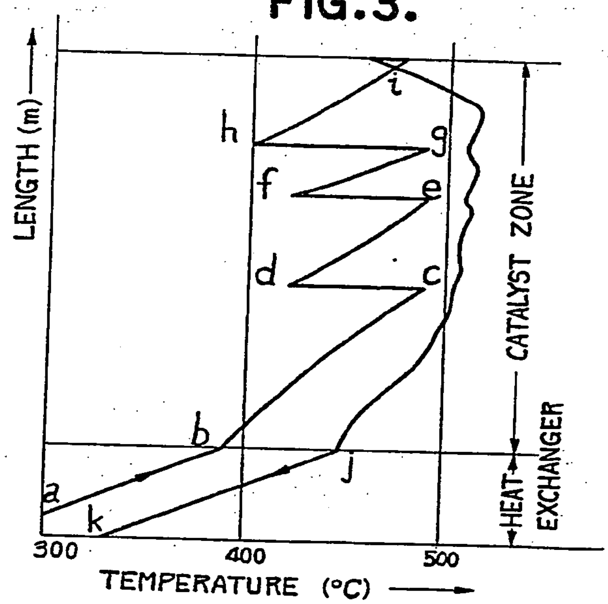


FIG.4.

I.

1089353 COMPLETE SPECIFICATION
2 SHEETS *This drawing is a reproduction of the Original on a reduced scale*
Sheets 1 & 2

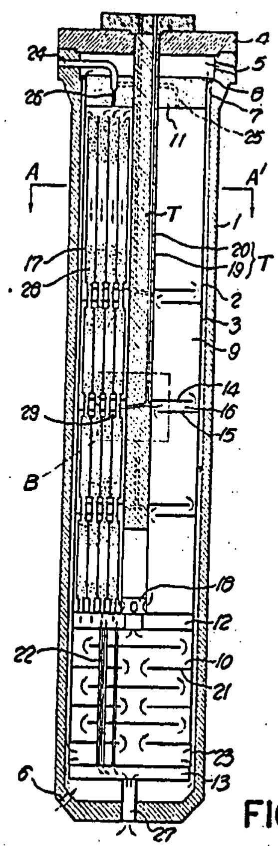


FIG.1.

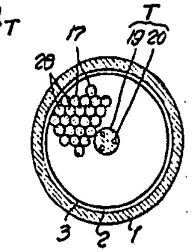


FIG.2.

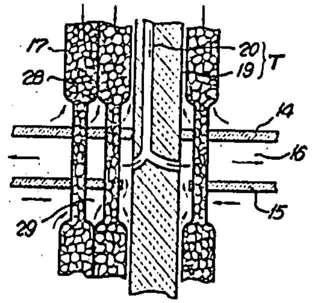


FIG.3.

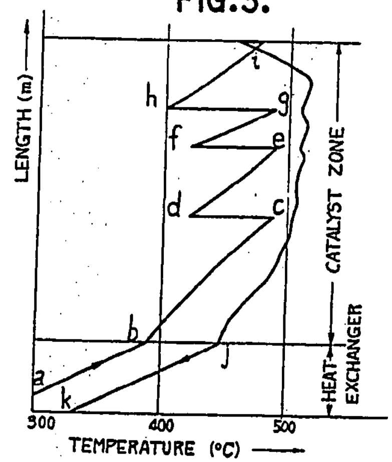


FIG.4.